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Exchange Interaction Model of Ferromagnetism

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Since $D_1^\dagger D_1 \geq 0$, it follows immediately from the minimax principle for the eigenvalues of a Hermitian operator that $H + D_1^\dagger D_1$ has at least as many positive eigenvalues as does H and at least as many nonnegative eigenvalues as H; since $P^{-1/2}$ is Hermitian positive definite, the number of positive eigenvalues as well as the number of zero eigenvalues of the operators $H + D_1^\dagger D_1$ and $P^{-1/2}(H + D_1^\dagger D_1)P^{-1/2}$ are identical. Therefore $P^{-1/2}(H + D_1^\dagger D_1)P^{-1/2}$ has at least J positive eigenvalues and at least $J + M$ nonnegative eigenvalues. Similarly, since $D_2^\dagger D_2 \geq 0$, $-P^{-1/2}(H + D_2^\dagger D_2)P^{-1/2}$ has at least J negative eigenvalues and at least $J + M$ nonpositive eigenvalues.

Theorem 5: Let N ($< n \equiv \dim E$) be the number of negative eigenvalues of H counted according to their multiplicity. Suppose the system (1) admits a Liapunov operator of the form $Lp(T)$, where $p(x)$ is a real polynomial in x (i.e., suppose the system is structurally stable). Then the degree of $p(x)$ need not exceed $4N + 1$, i.e., there exists a real polynomial $f(x)$ of degree less than or equal to $4N + 1$ such that $Lf(T)$ is a Liapunov operator.

Proof: Let N ($< n$) be the number of negative eigenvalues of H and let $\{\xi_k\}_1^{2n}$ be a complete L-canonical set of eigenvectors satisfying Eqs. (15) and (16) with real eigenvalues ω_k enumerated so that $\omega_1 \geq \omega_2 \geq \cdots \geq \omega_n$ and $\omega_{n+1} \geq \omega_{n+2} \geq \cdots \geq \omega_{2n}$. If 0 is not an eigenvalue of H, we conclude from Theorem 4, with $J = n - N$, that $\omega_{n+N+1} < 0 < \omega_{n-N}$. If 0 is an eigenvalue of H, then 0 is an eigenvalue of T [$H\eta = 0$ implies $T\begin{pmatrix}\eta\\0\end{pmatrix} = 0$]. By assumption, the system is structurally stable, so that the sets $Q_+ \equiv \{\omega_1, \ldots, \omega_n\}$ and $Q_- \equiv \{\omega_{n+1}, \ldots, \omega_{2n}\}$ are disjoint, and therefore either $0 \in Q_+$ or $0 \in Q_-$, but 0 cannot be in both. If $0 \in Q_+$, then we conclude from Theorem 4 (with $J + M = n - N$) that $\omega_{n+N+1} < 0 \leq \omega_{n-N}$, while if $0 \in Q_-$, Theorem 4 implies that $\omega_{n+N+1} \leq 0 < \omega_{n-N}$. Thus, in any case, $\omega_{n+N+1} < \omega_{n-N}$. Let r be the greatest integer $\leq n$ such that $\omega_r > \omega_{n+N+1}$, and let u be the least integer $\geq n + 1$ such that $\omega_u < \omega_r$. Then $n - N \leq r \leq n$, $n + 1 \leq u \leq n + N + 1$, $\omega_k < \omega_u$ for $r + 1 \leq k \leq n$, and $\omega_k > \omega_r$ for $n + 1 \leq k \leq u - 1$. Let $\omega_u < \rho < \omega_r$, and define $p_1(x) \equiv x - \rho$. Then $p_1(\omega_k)\gamma_k > 0$ for $1 \leq k \leq r$ and $u \leq k \leq 2n$. Let $\Omega_1^+, \ldots, \Omega_q^+$ denote the distinct elements of the set $\{\omega_k | r + 1 \leq k \leq n\}$ and let $\Omega_1^-, \ldots, \Omega_y^-$ denote the distinct elements of the set $\{\omega_k | n + 1 \leq k \leq u - 1\}$, so that $q \leq n - r \leq N$ and $y \leq u - 1 - n \leq N$. Since $\Omega_k^+ \in Q_+$ and $Q_+ \cap Q_- = \phi$, there exists $\epsilon > 0$ such that the q closed intervals $I_k^+ \equiv [\Omega_k^+ - \epsilon, \Omega_k^+ + \epsilon]$ are all disjoint and satisfy $I_k^+ \cap Q_- = \phi$, $k = 1, \ldots, q$. Similarly, there exists $\delta > 0$ such that the y closed intervals $I_k^- \equiv [\Omega_k^- - \delta, \Omega_k^- + \delta]$ are all disjoint and satisfy $I_k^- \cap Q_+ = \phi$, $k = 1, \ldots, y$. We define

$$p(x) \equiv p_1(x)\left[\prod_1^q (x - \Omega_k^+ + \epsilon)(x - \Omega_k^+ - \epsilon)\right] \times \left[\prod_1^y (x - \Omega_k^- + \delta)(x - \Omega_k^- - \delta)\right]. \quad (24)$$

Then by construction, Eq. (19) holds, so that $Lp(T) > 0$ and is therefore a Liapunov operator. The degree of $p(x)$ is given by $1 + 2q + 2y \leq 1 + 2N + 2N = 1 + 4N$, and the proof is complete.

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[1] E. M. Barston, J. Math. Phys. 12, 1116 (1971).
[2] E. M. Barston, J. Math. Phys. 8, 523 (1967).
[3] E. M. Barston, J. Math. Phys. 8, 1886 (1967).
[4] E. M. Barston, J. Math. Phys. 9, 2069 (1968).
[5] E. M. Barston, J. Math. Phys. 12, 1867 (1971).

Exchange Interaction Model of Ferromagnetism*

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The Schrödinger exchange operator for arbitrary spin has been used to form an interaction Hamiltonian for a nearest-neighbor model of ferromagnetism. Through use of the cluster expansion method and new group theoretic results in conjunction with the diagrammatic method, eight terms in the high temperature series for the zero-field partition function and the low-field susceptibility are obtained for arbitrary spin and general crystal lattice. Critical parameters are estimated from these series by means of various ratio tests and Padé approximants. For the cubic lattices the Curie temperature T_c and the critical index γ are given by

$$k_B T_C/J = 0.547(q - 1.6)(Y^{-1} + 0.21)$$

and

$$\gamma = 0.48 + 2.16\,Y^{-1} \quad \text{for } S > \tfrac{1}{2};$$
$$= 1.41 \pm 0.02 \quad \text{for } S = \tfrac{1}{2},$$

respectively, where $Y = 2S + 1$. Comparison of these results with those appropriate to the Heisenberg model as well as to experimental values is made. The concept of multipolar ordering is also discussed. It is shown that for the present model all of the $2S$ "independent" multipolar phase transitions are exactly degenerate with the usual dipolar transition.

I. INTRODUCTION

For the Heisenberg model of ferromagnetism, high temperature series for various thermodynamic quantities have been extensively studied and used to investigate critical properties of ferromagnetic systems. If we consider a ferromagnetic system contain-

ing N particles of spin S with isotropic nearest-neighbor exchange interactions, the Heisenberg Hamiltonian is given by

$$\mathcal{H} = -2J \sum_{<ij>} \mathbf{S}_i \cdot \mathbf{S}_j - g\mu H \sum_{i=1}^{N} S_{zi}, \tag{1}$$

where J is the nearest-neighbor exchange constant, $\mathbf{S}_i$ is the spin operator of an atom located at the lattice site labeled i, g is the gyromagnetic ratio, μ the Bohr magneton, H the z-directed external magnetic field, and S_{zi} is the z component of $\mathbf{S}_i$. The first term in the Hamiltonian represents the exchange energy and the summation is taken over all nearest-neighbor pairs of atoms, denoted $\langle ij \rangle$; the sum in the second term is over all atoms and is the Zeeman energy of the system.

The Heisenberg Hamiltonian linear in $\mathbf{S}_i \cdot \mathbf{S}_j$ which arises from a consideration of the Coulomb interaction together with the Pauli principle is in fact only the lowest-order significant term in a perturbation expansion which when carried further leads to terms nonlinear in $\mathbf{S}_i \cdot \mathbf{S}_j$.[1]

In order to study the properties of systems containing nonlinear terms $J_n(\mathbf{S}_i \cdot \mathbf{S}_j)^n$, in the Hamiltonian, the Schrödinger exchange operator has been used to form an interaction Hamiltonian for a nearest-neighbor model of ferromagnetism,[2,3]

$$\mathcal{H} = -J \sum_{<ij>} P_{ij} - g\mu H \sum_{i=1}^{N} S_{zi}. \tag{2}$$

Here P_{ij} is the Schrödinger exchange operator and is a polynomial of degree $2S$ in $\mathbf{S}_i \cdot \mathbf{S}_j$:

$$P_{ij} = \sum_{n=0}^{2S} A_n(S)(\mathbf{S}_i \cdot \mathbf{S}_j)^n, \qquad i \neq j. \tag{3}$$

The coefficients A_n are determined from the property that P_{ij} exchanges, or permutes, the spin coordinates of two atoms labeled i and j:

$$P_{ij} |m\rangle_i |m'\rangle_j = |m'\rangle_i |m\rangle_j \tag{4}$$

or

$$P_{ij} O(i,j) = O(j,i) P_{ij}, \tag{5}$$

where $O(i,j)$ is any operator which contains the spin operators of atoms i and j and $|m\rangle_i$ and $|m\rangle_j$ are eigenstates of S_{zi} and S_{zj}, respectively. Schrödinger[4] has explicitly shown that

$$P_{ij} = (-1)^{2S} \left(1 + \sum_{p=1}^{2S} \frac{(-1)^p}{(p!)^2} \prod_{q=1}^{p} [M - q(q-1)] \right), \tag{6}$$

where $M \doteq 2[S(S+1) + \mathbf{S}_i \cdot \mathbf{S}_j]$. The coefficients A_n typically have the values

$$\begin{aligned}
&S = \tfrac{1}{2}: \ A_0 = \tfrac{1}{2}, \quad A_1 = 2, \\
&S = 1: \ A_0 = -1, \quad A_1 = 1, \quad A_2 = 1, \\
&S = \tfrac{3}{2}: \ A_0 = -\tfrac{67}{32}, \quad A_1 = -\tfrac{9}{8}, \quad A_2 = \tfrac{11}{18}, \quad A_3 = \tfrac{2}{9}, \\
&S = 2: \ A_0 = -1, \quad A_1 = -\tfrac{5}{2}, \quad A_2 = -\tfrac{13}{16}, \quad A_3 = \tfrac{1}{6}, \quad A_4 = \tfrac{1}{36},
\end{aligned} \tag{7}$$

etc.

Inclusion of these special combinations of nonlinear terms in the Hamiltonian may not be realized in nature. However, a study of this model gives detailed information about what effect such nonlinear terms should have on the critical properties of the system.

Due to the permutation property of the Schrödinger exchange operator (5), the high temperature series can be extended further with less effort for the present Hamiltonian than for the Heisenberg Hamiltonian. For the case $S = \frac{1}{2}$, the present Hamiltonian is identical to that of the Heisenberg model; Baker *et al.*[5] obtained terms through T^{-9} for the close-packed lattices and T^{-10} for the loose-packed lattices for the zero-field partition function series and the low-field susceptibility series. For $S = 1$, Allan and Betts[2] obtained eight terms in these series for the face-centered cubic lattice. Such a large number of terms was obtained through the use of the cluster expansion method in conjunction with a technique making use of "Branching diagrams"; this is practicable only for the case of S equal to $\frac{1}{2}$ or 1. We have developed a new method which can be applied to the case of arbitrary spin directly and hence obtained eight terms in the high temperature series for general crystal lattices.

High temperature series expansions and the cluster expansion method are discussed in Secs. II and III, respectively; these ideas can be applied to any of the spin Hamiltonians usually studied. Sections IV and V contain group theoretical considerations and the diagrammatic method required to calculate the series coefficients for the present model. Details of the calculations are given in Sec. VI and the explicit series results are presented in Sec. VII. Several checking procedures on the results are considered in Sec. VIII. In Sec. IX these high temperature series are used to estimate various critical properties by means of ratio tests and the method of Padé approximants. The significance of the results as well as the concept of multipolar ordering is found in Sec. X.

II. HIGH TEMPERATURE SERIES EXPANSIONS

A. Preliminary Remarks

For any spin Hamiltonian $\mathcal{H}$ the high temperature series expansion method introduced by Kramers[6] and Opechowski[7] makes use of a result of the form

$$\begin{aligned}
Z &= \mathrm{tr} e^{-\beta \mathcal{H}} \\
&= \mathrm{tr}\mathbf{I}[1 - \beta \langle \mathcal{H} \rangle + (\beta^2/2!)\langle \mathcal{H}^2 \rangle \\
&\quad - (\beta^3/3!)\langle \mathcal{H}^3 \rangle + \cdots],
\end{aligned} \tag{8}$$

where Z is the partition function, $\beta = (k_B T)^{-1}$, k_B is Boltzmann's constant, $\langle \mathcal{H}^n \rangle = \mathrm{tr}\mathcal{H}^n / \mathrm{tr}\mathbf{I}$, and $\mathbf{I}$ is the unit matrix. Related thermodynamic functions can then be expressed as ascending series in powers of $1/T$ by evaluating the leading coefficients in the series for various crystal lattices. The first few terms of these series provide a good approximation to each thermodynamic quantity at high temperatures. Furthermore, extrapolations from such truncated expansion series are considered to be the most powerful theoretical approach yet developed for obtaining estimates of the various critical parameters.

B. Zero-Field Partition Function and Related Thermodynamic Functions

Since the various thermodynamic functions are re-

lated to the partition function by $\ln Z$, it is convenient to express the partition function in the form $\ln Z$. Moreover, since $\ln Z$ is an extensive quantity, considerable simplification can be made in the derivation of high temperature series by means of the cluster expansion method, as will be discussed in the next section. We introduce the following convenient notations: $Y = 2S + 1$, $X = S(S + 1)$, $\alpha = g\mu H/J$, $K = J/k_BT$, and

$$Q = \sum_{i=1}^{N} S_{zi}, \tag{9}$$

$$\mathcal{P} = \sum_{<ij>} P_{ij}, \quad \text{for the exchange interaction model,} \tag{10}$$

$$= 2\sum_{<ij>} \mathbf{S}_i \cdot \mathbf{S}_j, \quad \text{for the Heisenberg model,}$$

$$= 2\sum_{<ij>} S_{zi} S_{zj}, \quad \text{for the Ising model.}$$

Then,

$$\mathcal{H} = -J(\mathcal{P} + \alpha Q) = \mathcal{H}_0 - J\alpha Q. \tag{11}$$

For a system which consists of N particles of spin S, $\mathrm{tr} I = Y^N$. In zero external field,

$$Z = Y^N \left(1 + \sum_{n=1}^{\infty} \frac{K^n}{n!} \langle \mathcal{P}^n \rangle\right), \tag{12}$$

and

$$\ln Z = N \ln Y + \ln \left(1 + \sum_{n=1}^{\infty} \frac{K^n}{n!} \langle \mathcal{P}^n \rangle\right), \tag{13}$$

from which it follows on expanding the logarithm

$$\ln Z = N \ln Y + \sum_{n=1}^{\infty} e_n \frac{K^n}{n!}, \tag{14}$$

with

$$e_n = \sum \frac{(-1)^{\alpha_1+\alpha_2+\cdots+\alpha_p-1}(\alpha_1 + \alpha_2 + \cdots + \alpha_p - 1)!}{\alpha_1!\alpha_2!\ldots\alpha_p!(a_1!)^{\alpha_1}(a_2!)^{\alpha_2}\ldots(a_p!)^{\alpha_p}} \times \langle \mathcal{P}^{a_1} \rangle^{\alpha_1} \langle \mathcal{P}^{a_2} \rangle^{\alpha_2} \ldots \langle \mathcal{P}^{a_p} \rangle^{\alpha_p}. \tag{15}$$

The summation is taken over all partitions of the integer n, namely, all sets of positive integers $(a_1, a_2, \ldots, a_p; \alpha_1, \alpha_2, \ldots, \alpha_p)$ which satisfy the conditions

$$a_1\alpha_1 + a_2\alpha_2 + \cdots + a_p\alpha_p = n \tag{16}$$

and

$$a_1 < a_2 < \cdots < a_p. \tag{17}$$

Each thermodynamic function series can now be directly obtained from Eq. (14):

internal energy:
$$E = k_B T^2 \frac{\partial}{\partial T} (\ln Z)$$
$$= -J \frac{\partial}{\partial K} (\ln Z), \tag{18}$$

entropy:
$$S = k_B \frac{\partial}{\partial T} (T \ln Z)$$
$$= -k_B K^2 \frac{\partial}{\partial K} \frac{(\ln Z)}{K}, \tag{19}$$

specific heat:
$$C_v = k_B \frac{\partial}{\partial T} \left(T^2 \frac{\partial}{\partial T} \ln Z\right)$$
$$= k_B K^2 \frac{\partial^2}{\partial K^2} (\ln Z). \tag{20}$$

C. Low-Field Susceptibility

The low-field susceptibility is defined as

$$\chi = \lim_{H \to 0} k_B T \frac{\partial^2}{\partial H^2} \ln Z = \lim_{\alpha \to 0} k_B T \left(\frac{g\mu}{J}\right)^2 \frac{\partial^2}{\partial \alpha^2} \ln Z. \tag{21}$$

Since $\mathcal{P}$ and Q commute, $(\partial/\partial\alpha)(\mathcal{P} + \alpha Q)^n = n(\mathcal{P} + \alpha Q)^{n-1} Q$. It is then straightforward to show that

$$\chi = [(g\mu)^2/k_BT]\Delta(Q), \tag{22}$$

where $\Delta(Q) = \langle Q^2 \rangle_\beta - \langle Q \rangle_\beta^2$ is the zero-field thermal fluctuation of Q, and, for any operator A,

$$\langle A \rangle_\beta = \frac{\mathrm{tr} A e^{-\beta \mathcal{H}_0}}{\mathrm{tr} e^{-\beta \mathcal{H}_0}} = \sum_{n=0}^{\infty} \frac{K^n}{n!} \langle \mathcal{P}^n A \rangle \Big/ \sum_{n=0}^{\infty} \frac{K^n}{n!} \langle \mathcal{P}^n \rangle. \tag{23}$$

As a consequence of the fact that $\ln Z$ is an even function of the external field, $\mathrm{tr}\, \mathcal{P}^n Q = 0$ for all n; hence, $\langle Q \rangle_\beta = 0$, so that we can rewrite χ in the form

$$\chi = \frac{(g\mu)^2 X}{3k_BT} \left(N + \sum_{n=1}^{\infty} a_n \frac{K^n}{n!}\right). \tag{24}$$

It is easy to show that the coefficients a_n satisfy the recursion relation

$$a_n = \frac{3}{X} \langle \mathcal{P}^n Q^2 \rangle - \sum_{k=0}^{n-1} \frac{n!}{k!(n-k)!} a_k \langle \mathcal{P}^{n-k} \rangle, \tag{25}$$

with $a_0 = N$. In order to obtain the terms e_k and a_k, we must calculate the quantities $\langle \mathcal{P}^n \rangle$ and $\langle \mathcal{P}^n Q^2 \rangle$ for $n \leq k$. Here the matrices $\mathcal{P}$ and Q are of order $(2S + 1)^N$. For real crystals, $N \to \infty$, and direct computation of the required traces is impossible. However, there are two alternative methods of handling the calculation. In the next section we discuss the cluster expansion method, while the diagrammatic method is considered in Sec. V.

III. CLUSTER EXPANSION METHOD

A. Preliminary Remarks

The use of the cluster expansion method in deriving series expansions for magnetic systems was first suggested by Domb.[8] It was pointed out that high temperature series for extensive quantities for infinite lattices ($N \to \infty$) can be obtained simply by calculating the corresponding series for clusters of finite sites. One advantage of this method as compared to the diagrammatic method is that the number of configurations that one has to consider are considerably smaller than that required in the diagrammatic method. More important, however, is the fact that most of the calculations called for in the cluster expansion method can easily be done on a fast computer.

The general method of the cluster expansions has been developed by many authors and derived in a number of different ways. In this section we introduce a new proof of an essential theorem of this method (Theorem 1), which can be applied directly to the calculation of the high temperature series for any spin Hamiltonian.

B. General Concepts

A (linear) graph is a collection of points with lines joining certain pairs of points. If a subset of points

are joined successively by lines, the assembly of these lines is called a path connecting the initial and the final points. A graph is said to be connected if any two points in the graph are connected by a path. Otherwise, the graph is said to be disconnected. Clearly, any graph consists of connected graphs, and each connected graph is called a component. If the initial point and the final point of a path coincide, we speak of a cycle. A set of different cycles is said to be independent if none of the cycles can be made up of parts of other cycles. The maximum number of independent cycles in a graph is called the cyclomatic number of the graph. It is well known that for a connected graph

$$c = \ell - p + 1, \tag{26}$$

where c is the cyclomatic number, ℓ the number of lines, and p the number of points in a graph. In general, if we denote the number of connected components in a graph by n, then

$$c = l - p + n. \tag{27}$$

A connected graph is said to be closed if any point in the graph has at least two lines connected to it. Otherwise, it is said to be open.

If g_r and g_s are two graphs having no points in common, the union of these two graphs, denoted $g_r \cup g_s$, is the collection of all points and lines of g_r and g_s. A graph g is a subgraph of G if any point in g is a point in G and any line in g is a line in G. A graph G' is said to be isomorphic with G if there is a one-to-one correspondence between their points such that pairs of points are joined by lines in G' if and only if the corresponding pairs of points are joined in G. The lattice constant[9] of a graph g on a graph G is the number of subgraphs of G isomorphic with g, denoted $(g; G)$. $(g; G)$ is sometimes abbreviated as $[g]$ if G is not specified. Lattice constants of disconnected graphs can be expressed in terms of lattice constants of connected graphs. For example, consider the equation shown in Fig. 1. The first term on the right-hand side is of second order while the others are of first order in the lattice constants of connected graphs. In general, lattice constants of disconnected graphs having n connected components will consist of terms from the first order to the nth order in lattice constants of connected graphs.

C. New Derivation of the Cluster Expansions

Let $\phi(g)$ be any quantity associated with the graph g. ϕ is said to be extensive if, for any graphs g_r and g_s having no points in common,

$$\phi(g_r \cup g_s) = \phi(g_r) + \phi(g_s); \tag{28}$$

that is, the quantity ϕ of two graphs considered together is the sum of the quantities of the two graphs considered separately. The number of lines and the number of points in a graph are obviously extensive. $(g; G)$ is also an extensive quantity of G, i.e.,

$$(g; g_r \cup g_s) = (g; g_r) + (g; g_s). \tag{29}$$

Suppose that a graph G consists of the number π_i of connected graphs g_i, $i = 1, 2, 3, \cdots$. Let $\phi(G; t)$ be an extensive quantity of G and t be a set of parameters independent of graphs. By the extensive property of ϕ,

$$\phi(G; t) = \sum_i \phi(g_i; t)\pi_i. \tag{30}$$

Using the extensive property of the lattice constant as expressed by Eq. (29), set $g = g_j$ for $j = 1, 2, 3, \cdots$. We then have a set of linear equations

$$(g_j; G) = \sum_i (g_j; g_i)\,\pi_i, \quad j = 1, 2, 3, \cdots. \tag{31}$$

If the graphs are labeled in the graph dictionary order such that

$$l_i \leq l_j, \quad \text{for } i < j, \tag{32}$$

where l_i and l_j are the numbers of lines of graphs g_i and g_j, respectively, it is then obvious that

$$\begin{aligned}(g_j; g_i) &= 0, \quad \text{for } j > i, \\ &= 1, \quad \text{for } j = i.\end{aligned} \tag{33}$$

Define a matrix A with elements $A_{mn} = (g_m; g_n)$. Equation (33) then means that A is a triangular matrix with the lower triangular elements equal to zero. Furthermore, all the diagonal elements are unity. Therefore, A is nonsingular and its inverse exists. From Eq. (31), we get

$$\pi_i = \sum_j (A^{-1})_{ij}(g_j; G), \quad i = 1, 2, 3, \ldots, \tag{34}$$

where $(A^{-1})_{ij}$ are elements of the inverse matrix of A. Substituting Eq. (34) into Eq. (30) yields

$$\begin{aligned}\phi(G; t) &= \sum_i \sum_j \phi(g_i; t)(A^{-1})_{ij}(g_j; G) \\ &= \sum_j F_j(t)(g_j; G),\end{aligned} \tag{35}$$

where $F_j(t) = \sum_i (g_i; t)(A^{-1})_{ij}$ is independent of G. Since G may be any graph, let $G = g_i$. Equation (35) yields

$$\phi(g_i; t) = \sum_j F_j(t)(g_j; g_i). \tag{36}$$

On substituting Eq. (33) into Eq. (36) and rearranging terms, we obtain an important theorem formulated by Sykes *et al.*[10]

Theorem 1: If $\phi(G; t)$ satisfies the extensive property, then ϕ can be expressed by Eq. (35) in which $F_j(t)$ are given by the recursion formula

$$F_j(t) = \phi(g_j; t) - \sum_{i=1}^{j-1} (g_i; g_j)F_i(t) \tag{37}$$

and

$$F_1(t) = \phi(g_1; t). \tag{38}$$

D. Application to Magnetic Systems

In a nearest-neighbor model of any of the spin Hamiltonians, if spin sites are represented by points and interactions between nearest-neighbor sites are repre-

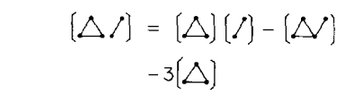

FIG. 1. Example of expressing the lattice constant of a disconnected graph in terms of lattice constants of connected graphs.

sented by lines joining the corresponding pairs of points, then systems represented by graphs isomorphic to each other will have the same physical properties and systems which consist of a number of independent subsystems will be represented by disconnected graphs. Let $\phi(G)$ be any quantity of a magnetic system represented by a graph G. It is clear then that $\ln Z(G)$ and $\chi(G)$ satisfy Eq. (28) since $\ln Z$ and χ are extensive thermodynamic quantities. We can then use Theorem 1 to calculate $\ln Z(G; t)$ and $\chi(G; t)$ for any cluster or crystal lattice G and for a set of parameters t, such as temperature T, spin value S, external magnetic field H, exchange constant J, etc. In high temperature series expansions, we express F_j [in Eq. (35)] in powers of $K(= J/k_B T)$

$$F_j = \sum_{n\ 0}^{\infty} f_n(g_j) K^n. \tag{39}$$

Theorem 1 is useful because in most cases $f_n(g_j) = 0$ for $n < l_j$. This will be explicitly shown in Sec. V for the present Hamiltonian when $\phi = \ln Z$ or χ. Therefore, if we want to obtain series up to the kth power in K for $\phi(G)$, we need only calculate $\phi(g_j)$ and hence F_j for connected graphs having up to k lines. Thus, instead of considering infinite lattices, we have reduced the problem to clusters of finite size.

IV. APPLICATION OF GROUP THEORY

A. Symmetric Group

A rearrangement of the order of N symbols is called a permutation. There are $N!$ number of possible permutations on N symbols. The set of all these permutations form a group called the symmetric group of degree N, denoted S_N. Each elements in S_N can be written as a product of independent cycles. For example, the permutation by which $\{a, b, c, d, e, f, g\}$ is replaced by $\{b, f, c, e, d, a, g\}$ can be written as $(abf)(c)(de)(g)$. Each ( ) is called a cycle, and the number of symbols in ( ) is the order of the cycle. In writing a permutation as the product of cycles, cycles of order 1 need not be mentioned. Furthermore, the sequence of appearance of the cycles as well as the first symbol in each cycle is arbitrary.

Cycles having no symbols in common are said to be independent. A cycle of order 2 is called an interchange. Any cycle of higher order can be expressed as a product of interchanges (having symbols in common). For example,

$$(abc\cdots de) = (ae)(ad)\cdots(ac)(ab). \tag{40}$$

Permutations which are products of an even number of interchanges are called even permutations. Otherwise, they are called odd permutations. It is straightforward to show that

$$(fg)(fa\cdots bgc\cdots d) = (fa\cdots b)(gc\cdots d) \tag{41}$$

and

$$(fg)(fa\cdots b)(gc\cdots d) = (fa\cdots bgc\cdots d). \tag{42}$$

Equations (41) and (42) imply that for any group element, say P, when multiplied by an interchange (fg), $(fg)P$ has one more cycle than P if f, g belong to the same cycle in P and has one less cycle than P if f, g belong to different independent cycles in P.

Elements in S_N are divided into classes. Elements which have the same cycle structure are transforms of one another by elements in S_N and belong to the same class. Elements which belong to the class $(a_1^{\alpha_1} a_2^{\alpha_2} \cdots a_p^{\alpha_p})$ have α_1 cycles of order a_1, α_2 cycles of order $a_2, \ldots$, and α_p cycles of order a_p. The number of classes in S_N is equal to the number of partitions of the integer N, and each class is labeled by a partition of N. The number of group elements in a class is called the order of the class. For the class $k = (a_1^{\alpha_1} a_2^{\alpha_2} \cdots a_p^{\alpha_p})$, its order h_k is

$$h_k = N!/\alpha_1!\alpha_2! \cdots \alpha_p! a_1^{\alpha_1} a_2^{\alpha_2} \cdots a_p^{\alpha_p}. \tag{43}$$

Associated with S_N there is a finite number of inequivalent representations, called irreducible representations. Each irreducible representation is also labeled by a partition of N. Since elements of a class are transforms of one another, their matrices in any representation, say ν, have the same trace. This value is referred to as the character of the class k in the representation ν, denoted $\chi_k^{(\nu)}$.

Below we summarize some group theoretic results which will be useful in deriving high temperature series for the present Hamiltonian.

For the irreducible representations ν and ν', we have the orthogonality relations

$$\sum_k \frac{h_k}{h} \chi_k^{(\nu)} \chi_k^{(\nu')} = \delta_{\nu\nu'}, \tag{44}$$

and

$$\sum_\nu \frac{h_k}{h} \chi_k^{(\nu)} \chi_{k'}^{(\nu)} = \delta_{kk'}, \tag{45}$$

where h_k is the order of the class k, $h = N!$ is the order of S_N, and the summations $\sum_k$ and $\sum_\nu$ are taken over all classes k and all irreducible representations ν, respectively.

Let $P^{(\Gamma)}$ be any matrix representation of an element P in S_N. $P^{(\Gamma)}$ can be resolved into a direct sum of n_ν number of ν irreducible representations, i.e., the matrix $P^{(\Gamma)}$ now takes the form of a series of blocks, the irreducible representations, placed along the principal diagonal, which can be written as

$$P^{(\Gamma)} = \sum_\nu n_\nu P^{(\nu)}. \tag{46}$$

From Eq. (44), we get

$$n_\nu = \sum_k \frac{h_k}{h} \chi_k^{(\nu)} \chi_k^{(\Gamma)}. \tag{47}$$

For two irreducible representations labeled by a pair of conjugate partitions ν and $\tilde{\nu}$, we have

$$\chi_k^{(\nu)} = \pm \chi_k^{(\tilde{\nu})}, \tag{48}$$

with the plus sign applying for even classes and the minus sign for odd classes of permutations.

If we sum the matrices of an irreducible representation ν for all elements of a class k, we obtain a multiple of the unit matrix:

$$\sum_{P \in k} P^{(\nu)} = [(h_k \chi_k^{(\nu)})/\chi_{1^N}^{(\nu)}] \mathrm{I}, \tag{49}$$

where $\chi_{1^N}^{(\nu)}$ is the matrix dimension of the irreducible representation ν.

Using these equations, we now prove several useful new theorems.

B. New Theorems

Theorem 2: If R is any matrix which commutes with all elements of S_N in a matrix representation Γ, then

$$\mathrm{tr}[(\mathcal{P}^{(\Gamma)})^n R] = \sum_\nu \sum_k \mathrm{tr}(\mathcal{P}^{(\nu)})^n \frac{h_k}{h} \chi_k^{(\nu)} \mathrm{tr}[P_k^{(\Gamma)} R], \tag{50}$$

where $\mathcal{P}^{(\Gamma)}$ and $\mathcal{P}^{(\nu)}$ are sums of elements in S_N in the Γ and the ν representations, respectively, P_k is any element in the class k, and the summations are taken over all irreducible representations ν and all classes k.

Proof: The condition that R commutes with all elements of S_N in a matrix representation Γ and the fact that elements of a class are transforms of one another imply that $\mathrm{tr}P^{(\Gamma)}R$ have the same value for all elements P which belong to the same class. Since products of elements in S_N are also elements in S_N, $\mathcal{P}^n$ as well as $\mathcal{P}$ is a sum of elements in S_N. It is then sufficient to show that for any element, say P, in S_N

$$\mathrm{tr}P^{(\Gamma)}R = \sum_\nu \sum_k \mathrm{tr}P^{(\nu)} \frac{h_k}{h} \chi_k^{(\nu)} \mathrm{tr}P_k^{(\Gamma)} R. \tag{51}$$

Let P belong to the class k'; $\mathrm{tr}P^{(\nu)} = \chi_{k'}^{(\nu)}$. By Eq. (45)

$$\begin{aligned} &\sum_\nu \sum_k \chi_{k'}^{(\nu)} \frac{h_k}{h} \chi_k^{(\nu)} \mathrm{tr}P_k^{(\Gamma)} R \\ &\quad = \sum_k \delta_{kk'} \mathrm{tr}P_k^{(\Gamma)} R \\ &\quad = \mathrm{tr}P_{k'}^{(\Gamma)} R. \end{aligned} \tag{52}$$

Equation (51) and hence Eq. (50) then follow.

Theorem 3: If ν and $\tilde{\nu}$ are two irreducible representations labeled by a pair of conjugate partitions of N and $\mathcal{P}$ is a sum of odd permutations in S_N, then

$$\mathrm{tr}(\mathcal{P}^{(\nu)})^n = (-1)^n \mathrm{tr}(\mathcal{P}^{(\tilde{\nu})})^n. \tag{53}$$

Proof: Since a product of n odd permutations is an even permutation if n is an even number and is an odd permutation if n is odd, Eq. (53) follows from Eq. (48).

C. The $(2S+1)^N$-Dimensional Representation of S_N

For a system containing N particles of spin S, it is clear that matrices of the Schrödinger exchange operators P_{ij} and their products form a $(2S+1)^N$-dimensional representation of S_N. It is convenient to choose the basis of the $(2S+1)^N$-dimensional vector space as the eigenstates of the z component of the spins of the N particles, $|m_1\rangle |m_2\rangle \cdots |m_N\rangle$. In this section we restrict our attention to this representation. Unless otherwise specified, matrix representation of operators will be in this representation. We first prove the following results:

$$\begin{aligned} \mathrm{tr}(ijk\cdots l) &= \sum_{m_i} \sum_{m_j} \cdots \sum_{m_k} \sum_{m_l} (\delta_{m_i,m_j} \delta_{m_i,m_k} \cdots \delta_{m_i,m_l}) \\ &= \sum_{m_i} (1) = Y. \end{aligned} \tag{54}$$

All sums in this equation, and those following directly after, range from $-S$ to $+S$. Similarly,

$$\begin{aligned} \mathrm{tr}S_{zi}^n (ijk\cdots l) &= \sum_{m_i} \sum_{m_j} \cdots \sum_{m_k} \sum_{m_l} (m_i^n \delta_{m_i,m_j} \delta_{m_i,m_k} \cdots \delta_{m_i,m_l}) \\ &= \sum_{m_i} (m_i^n) = YW_n, \end{aligned} \tag{55}$$

where

$$W_n = Y^{-1} \sum_m (m^n), \tag{56}$$

and

$$\begin{aligned} &\mathrm{tr}S_{zi}^n S_{zj}^n (ik\cdots jl\cdots) \\ &\quad = \sum_{m_i} \sum_{m_k} \cdots \sum_{m_j} \sum_{m_l} \cdots (m_i^n m_j^n \delta_{m_i,m_k} \cdots \delta_{m_i,m_j} \delta_{m_i,m_l} \cdots) \\ &\quad = \sum_{m_i} (m_i^{2n}) = YW_{2n}. \end{aligned} \tag{57}$$

Let us express any element in S_N, say P, as a product of independent cycles:

$$P = (abc\cdots d)(ijk\cdots l)\cdots(xy\cdots z). \tag{58}$$

P may be considered as a direct product of each cycle,

$$P = (abc\cdots d) \times (ijk\cdots l) \times \cdots \times (xy\cdots z). \tag{59}$$

By the trace property of the direct product,

$$\mathrm{tr}(A \times B) = (\mathrm{tr}A)(\mathrm{tr}B), \tag{60}$$

it follows from Eqs. (54), (55), and (57) that

$$\mathrm{tr}S_{zi}^n P = W_n \mathrm{tr}P, \tag{61}$$

and

$$\begin{aligned} \mathrm{tr}S_{zi}^n S_{zj}^n P &= W_{2n} \mathrm{tr}P, && \text{if } i=j \text{ or } i,j \text{ belong to the same cycle in } P, \\ &= W_n^2 \mathrm{tr}P, && \text{if } i,j \text{ belong to different independent cycles in } P. \end{aligned} \tag{62}$$

For the case $n=1$, $W_1 = 0$ and $W_2 = X/3$. If the element P belongs to the class $k = (a_1^{\alpha_1} a_2^{\alpha_2} \cdots a_p^{\alpha_p})$, then, from Eqs. (54), (60), and (62),

$$\mathrm{tr}P_k = Y^{\alpha_1+\alpha_2+\cdots+\alpha_p} \tag{63}$$

and

$$\begin{aligned} \mathrm{tr}P_k Q^2 &= \sum_i \sum_j \mathrm{tr}S_{zi} S_{zj} P_k \\ &= (X/3)(\alpha_1 a_1^2 + \alpha_2 a_2^2 + \cdots + \alpha_p a_p^2) Y^{\alpha_1+\alpha_2+\cdots+\alpha_p}, \end{aligned} \tag{64}$$

where the summations are from 1 to N.

It also follows from Eqs. (41), (42) and Eqs. (54), (60) that

$$\begin{aligned} \mathrm{tr}P_{ij} P &= Y\mathrm{tr}P, && \text{if } i,j \text{ belong to the same cycle in } P, \\ &= Y^{-1}\mathrm{tr}P, && \text{if } i,j \text{ belong to different independent cycles in } P. \end{aligned} \tag{65}$$

V. DIAGRAMMATIC METHOD

A. Preliminary Remarks

The diagrammatic method has been extensively used in deriving high temperature series for various spin Hamiltonians. In this section, the procedure previously used for the Heisenberg Hamiltonian by Rushbrooke and Wood[11] will be modified for the exchange interaction Hamiltonian. The labor of evaluating series

coefficients by this method is much greater than the labor involved in the previously described cluster expansion method. However, there are a number of important results that can be directly proved by the diagrammatic technique which are not at all obvious from a consideration of the cluster expansion method.

B. Zero-Field Partition Function

For the exchange interaction model $\mathcal{P}^n$ in Eq. (12) is a sum of products ΠP_{ij} and each product contains n factors P_{ij}. There is a correspondence between products in $\mathcal{P}^n$ and diagrams of n lines on the lattice. For each of the n factors P_{ij} in the product, when we draw a straight line connecting lattice sites i and j, we obtain a diagram of n lines. The diagrams may be connected or disconnected, and may have more than one line joining a pair of points.

Following Rushbrooke and Wood,[11] we can write

$$\langle \mathcal{P}^n \rangle = \sum_{i=n} [D_i]\langle D_i \rangle, \tag{66}$$

where $\sum_{i=n}$ sums over all diagrams D_i of n lines. $[D_i]$ is the number of times that the diagram D_i will occur on the lattice. $\langle D_i \rangle$ is the weighting factor, or the contribution of the diagram D_i to $\langle \mathcal{P}^n \rangle$. For a diagram D having n lines between p points, following Rushbrooke and Wood,

$$\langle D \rangle = Y^{-p} \sum_{\text{perm}} \text{tr}(\,)(\,)\cdots(\,), \tag{67}$$

where each bracket is a Schrödinger exchange operator P_{ij} which corresponds to the line joining points i and j in the diagram D and $\sum_{\text{perm}}$ sums over all different permutations in the order of appearence of the brackets.

The occurrence factor of a diagram on a lattice has the same meaning as the lattice constant of a graph. The only difference is that for graphs we speak of diagrams which have at most one line connecting a pair of points. Some typical examples of the relation between the occurrence factors of diagrams and lattice constants of graphs are shown in Fig. 2. As mentioned in Sec. III, lattice constants of disconnected graphs can be expressed in terms of lattice constants of connected graphs. Let $\Lambda_i\Phi$ signify that part of Φ which is of ith order in the lattice constants of connected graphs. We can then write

$$\langle \mathcal{P}^n \rangle = \Lambda_1\langle \mathcal{P}^n \rangle + \Lambda_2\langle \mathcal{P}^n \rangle + \cdots. \tag{68}$$

As shown in the section on the cluster expansion method, Eq. (35), any extensive quantity, such as $\ln Z$ or χ is of first order in lattice constants of connected graphs. Hence by Eq. (12),

$$\ln Z = N \ln Y + \sum_{n=1}^{\infty} \frac{K^n}{n!} \Lambda_1\langle \mathcal{P}^n \rangle. \tag{69}$$

Those parts of higher order in lattice constants of connected graphs will cancel exactly when transformed from Z to $\ln Z$. Comparing Eqs. (14) and (69), we have

$$e_n = \Lambda_1\langle \mathcal{P}^n \rangle = \sum_{i=n} \Lambda_1[D_i]\langle D_i \rangle. \tag{70}$$

In the limit $N \to \infty$, lattice constants of connected graphs will be directly proportional to N. Λ_1 then means nothing more than "the part proportional to N."

C. Low-Field Susceptibility

When a finite external magnetic field is applied, the partition function can be written in the form

$$\ln Z = N \ln Y + \sum_{n=1}^{\infty} \frac{K^n}{n!} \Lambda_1\langle (\mathcal{P} + \alpha Q)^n \rangle. \tag{71}$$

Since $\mathcal{P}$ and Q commute and $\langle \mathcal{P}^n Q \rangle = 0$, it is easy to show that

$$\begin{aligned} \chi &= \lim_{H\to 0} k_B T \frac{\partial^2}{\partial H^2} \ln Z \\ &= \frac{g^2\mu^2}{k_B T} \sum_{n=0}^{\infty} \frac{K^n}{n!} \Lambda_1\langle \mathcal{P}^n Q^2 \rangle. \end{aligned} \tag{72}$$

For $n = 0$, $\langle Q^2 \rangle = NX/3$. Comparing Eqs. (24) and (72) yields

$$a_n = (3/X)\Lambda_1\langle \mathcal{P}^n Q^2 \rangle. \tag{73}$$

The situation in calculating $\langle \mathcal{P}^n Q^2 \rangle$ is similar to that involved in $\langle \mathcal{P}^n \rangle$. We again have a correspondence between terms ΠP_{ij} produced on expanding $\mathcal{P}^n$ and diagrams of n lines on the lattice, but now, besides the n lines, a diagram will contain two crosses. These are denoted +, and are placed on those sites from which we have extracted terms like $S_{zi}S_{zj}$ from Q^2. When two crosses coincide, we speak of a double cross, denoted *.

There are five kinds of ways in which two crosses may be added to a diagram D:

(a) D^*, a double cross superposed on D,

(b) D^{++}, two crosses superposed on different sites of D,

(c) D^{+} +, one cross superposed on D, another not on D,

(d) D *, a double cross not superposed on D,

(e) D + +, two separated crosses not superposed on D.

Since $\langle S_{zi} \rangle = 0$ and $\langle S_{zi}^2 \rangle = X/3$, by the trace property of the direct product, we immediately find that diagrams (c) and (e) have zero contributions to $\langle \mathcal{P}^n Q^2 \rangle$ and that diagram (d) contributes an amount $(X/3)\langle D \rangle$ to $\langle \mathcal{P}^n Q^2 \rangle$. Let p be the number of points contained in the diagram D. Then we have that the occurrence factor, for diagram (d), $[D\,*] = [D][\,\cdot\,] - p[D]$. Therefore, as far as terms of first order in lattice constants of connected graphs are concerned, it is equivalent to say that the diagram (d) has occurrence factor $[D]$ associated with the weighting factor $-(pX/3)\langle D \rangle$.

For diagram (a) the double cross can be superposed on any of the p points. For diagram (b) there are $p(p-1)$ ways to superpose the two crosses on D. If

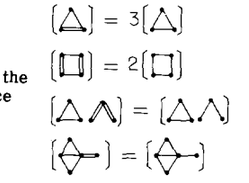

FIG. 2. Examples of the relation between the occurrence factors of diagrams and lattice constants of graphs.

we sum up these p^2 diagrams, with occurrence factor $[D]$, their contributions to $\langle \mathcal{P}^n Q^2\rangle$, denoted $\langle \overline{DQ}^2\rangle$, will be

$$\langle \overline{DQ}^2\rangle = Y^{-p} \sum_{\text{perm}} \text{tr}(\ )(\ )\cdots(\ )(\sum_{i=1}^{p} S_{zi})^2. \tag{74}$$

Therefore, including all five kinds of diagrams, with occurrence factor $[D]$, the weighting factor of the diagram D to $\Lambda_1\langle \mathcal{P}^n Q^2\rangle$, denoted $\langle DQ^2\rangle$, will be

$$\langle DQ^2\rangle = \langle \overline{DQ}^2\rangle - (pX/3)\langle D\rangle. \tag{75}$$

Hence

$$a_n = (3/X)\sum_{l_i=n} \Lambda_1[D_i][\langle \overline{D_i Q}^2\rangle - p_i(X/3)\langle D_i\rangle]. \tag{76}$$

where p_i is the number of points in D_i. By rewriting the products ΠP_{ij} as products of independent cycles, the traces in Eqs. (67) and (74) can be obtained from Eqs. (63) and (64). $\langle D_i\rangle$ and $\langle D_i Q^2\rangle$ are then determined. Note that for the present Hamiltonian these five kinds of diagrams can be considered together and the calculation of the coefficients a_n is greatly simplified. For the Heisenberg Hamiltonian, weighting factors of these five kinds of diagrams have to be considered separately.

D. Further Theorems

If we express $\Lambda_1[D_i]$ in terms of lattice constants of connected graphs, denoted $[g_i]$, we can rewrite e_n and a_n as follows:

$$e_n = \sum_i [g_i] e_n(g_i) \tag{77}$$

and

$$a_n = \sum_i [g_i] a_n(g_i), \tag{78}$$

where $\sum_i$ sums over all connected graphs g_i. We then prove the following:

Theorem 4:

$$e_n(g_i) = a_n(g_i) = 0, \quad \text{if } n < \ell_i. \tag{79}$$

Proof: This is obvious since those diagrams with occurrence factor containing $[g_i]$ must have ℓ_i lines or more, and they contribute to e_n and a_n for $n \geq \ell_i$.

Theorem 5: If we define cycles of diagrams similar to those for graphs presented in Sec. III, then for diagrams containing n lines and c independent cycles their contributions to e_n and a_n contain terms Y^{-n+2k}, where k ranges from 0 to c, but $2k \geq n$.

Proof: As mentioned in Sec. IV, for any group element P, $(fg)P$ has one more cycle than P if f, g be long to the same cycle in P and has one less than P if f, g belong to different independent cycles in P. Assume that the diagram contains p points. The identity element will have p different independent cycles, namely, (1) (2) $\cdots$ (p). Consider one of the products

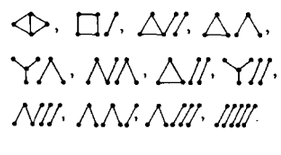

FIG. 3. Let the first graph be g_i. All diagrams of five lines with occurrence factors containing $[g_i]$ are shown. This illustrates the fact that diagrams of ℓ_i lines with occurrence factors containing $[g_i]$ consist of subgraphs of g_i.

of n factors (ij) occurring in Eqs. (67) and (74), and multiply successively to the identity element, first, the nth factor, then the $(n-1)$th factor, etc., and finally the first factor. Since each multiplication either decrease or increase the number of different independent cycles by 1, the resultant product will contain $p - n + 2k$ different independent cycles. Here k is the number of times that i, j happen to occur in the same cycle in the product which is to be multiplied by (ij). This can happen only when i, j are joined by paths other than the line ij, or ij must be a line of a cycle. Therefore, $k \leq c$. Also, the number of independent cycles may not be greater than p, $2k \leq n$. Equation (63) says that for any permutation containing t different independent cycles its trace in the $(2S+1)^p$-dimensional matrix representation is Y^t. Theorem 5 then follows from a consideration of Eqs. (63), (67), (70) and Eqs. (64), (74), and (76).

A consequence of this theorem is that $e_n(g_i)$ and $a_n(g_i)$ contain terms $Y^{-n}, Y^{-n+2}, \cdots$, which we can write as

$$e_n(g_i) = e_n^{(n)}(g_i)Y^{-n} + e_n^{(n-2)}(g_i)Y^{-n+2} + \cdots + e_n^{(1)}(g_i)Y^{-1} \quad [\text{or } e_n^{(0)}(g_i)] \tag{80}$$

and

$$a_n(g_i) = a_n^{(n)}(g_i)Y^{-n} + a_n^{(n-2)}(g_i)Y^{-n+2} + \cdots + a_n^{(1)}(g_i)Y^{-1} \quad [\text{or } a_n^{(0)}(g_i)], \tag{81}$$

Theorem 6:

$$e_n^{(-n+2k)}(g_i) = a_n^{(-n+2k)}(g_i) = 0, \quad \text{for } k > c_i + n - \ell_i, \tag{82}$$

where c_i is the number of independent cycles and ℓ_i is the number of lines in the connected graph g_i.

Proof: Consider first the case that $n = \ell_i$. Diagrams of ℓ_i lines which have an occurrence factor containing $[g_i]$ are those which consist of subgraphs of g_i, and hence cannot have more cycles than g_i. For example, let g_i be the connected graph in Fig. 3. Other diagrams of five lines with occurrence factor containing $[g_i]$ are those disconnected graphs shown in the figure. Hence from Theorem 5, Theorem 6 is proved for the case $n = \ell_i$. When $n > \ell_i$, we can superpose the additional $n - \ell_i$ lines on g_i. Each line superposed on g_i is equivalent to forming an additional cycle in the resultant diagram, and the maximum number of independent cycles will be $c_i + n - \ell_i$ in some of the n diagrams which have occurrence factors containing $[g_i]$. This then completes the proof of Theorem 6.

Theorem 7:

$$a_n^{(0)}(g_i) = 0 \quad \text{for all } g_i. \tag{83}$$

Proof: Consider a diagram D of p points and n lines which has occurrence factor containing $[g_i]$. Those products ΠP_{ij} in Eqs. (67) and (74) which contribute to $a_n^{(0)}$ must be equal to the identity operator. Since $\text{tr}\mathbb{I}Q^2 = pX/3$, Eq. (83) follows from Eq. (75).

Theorem 8:

$$e_n^{(1)}(g_i) = \tfrac{1}{2} a_n^{(1)}(g_i) \quad \text{for all } g_i. \tag{84}$$

Proof: Those products in Eqs. (67) and (74) which contribute to $e_n^{(1)}$ and $a_n^{(1)}$ must contain $p-1$ cycles, or belong to the class $1^{p-2}2$. From Eqs. (63) and (64), for permutations P belonging to the class $1^{p-2}2$,

$$\mathrm{tr}P\left(\sum_{i=1}^{p} S_{zi}\right)^2 = \frac{X}{3}(p+2)Y^{p-1} \tag{85}$$

and

$$\mathrm{tr}P = Y^{p-1}. \tag{86}$$

From Eqs. (67), (74), and (75), the contributions of these products to $\langle D\rangle$ and $(3/X)\langle DQ^2\rangle$ will be Y^{-1} and $2Y^{-1}$, respectively, Theorem 8 then follows.

VI. CALCULATIONS OF SERIES COEFFICIENTS

A. Clusters With Up to Seven Sites

From the discussion in Sec. III we know that in order to obtain the high temperature series for general lattices up to the seventh power in $K(=J/k_BT)$, we must calculate the corresponding series for clusters having up to seven lines. For computational convenience we group these clusters into two categories, those having up to seven points and those containing eight points. Consider first clusters with up to seven points and seven lines. They are labeled in the graph dictionary order such that Eq. (33) is satisfied.[12]

To calculate the coefficients in the zero-field partition function and the low-field susceptibility series for finite clusters we make use of Eqs. (15) and (25), in which the quantities $\mathrm{tr}\mathcal{P}^n$ and $\mathrm{tr}\mathcal{P}^nQ^2$ for $n \leq 7$ are obtained from Theorem 2 by setting R equal to the unit matrix and Q^2, respectively. Now $\mathcal{P}^{(\Gamma)}$ in Eq. (50) is a sum of Schrödinger exchange operators which correspond to the lines in each cluster. h_k and $\chi_k^{(\nu)}$ are available in a number of texts which deal specifically with the symmetric group.[13] The values of $\mathrm{tr}P_k$ and $\mathrm{tr}P_kQ^2$ can be calculated by Eqs. (63) and (64). Corresponding to $\mathcal{P}^{(\Gamma)}, \mathcal{P}^{(\nu)}$ is a sum of interchanges in the irreducible representation ν. The explicit form of the permutation matrices in any irreducible representation can be obtained by the technique introduced by Yamanouchi.[14]

For two conjugate representations $\mathrm{tr}\mathcal{P}^n$ are related to each other by Eq. (53) and hence only one of the traces need by calculated. The quantities $\mathrm{tr}(\mathcal{P}^{(\nu)})^n$ were calculated on a computer for all required clusters with up to seven points. The size of the greatest matrix involved in the calculation is of dimension 35×35.

B. Clusters Containing Eight Sites

For clusters with seven lines and eight sites, we can still evaluate the high temperature series following the procedure just described. However, for the symmetric group of degree eight some of the irreducible representations are of dimension 70×70 and the trace calculation on a computer would have been too expensive for us. From Eq. (26) we have that clusters with seven lines and eight points contain no cycles. It then follows immediately from Theorem 4 that for these clusters $e_n(g_i) = a_n(g_i) = 0$ for $n \leq 6$ and from Theorem 6 that $e_7(g_i)$ and $a_7(g_i)$ can be written in the form

$$e_7(g_i) = e_7^{(7)}(g_i)Y^{-7} \tag{87}$$

and

$$a_7(g_i) = a_7^{(7)}(g_i)Y^{-7}. \tag{88}$$

For $S = \frac{1}{2}$ the exchange interaction Hamiltonian is identical to the Heisenberg Hamiltonian, and the high temperature series for these two models are the same. It is known that for the Heisenberg model graphs with n lines containing no cycles will not contribute to e_n and the only graph with n lines containing no cycles which contributes to a_n is the simple chain.[15] Therefore, $e_7(g_i) = a_7(g_i) = 0$ for all clusters of eight points with the exception that for the simple chain $a_7(g_i) = 10\,080Y^{-7}$. This value was obtained from Eq. (88) together with the results of Domb and Wood[15]; for the case of spin $\frac{1}{2}$ and for the simple chain of eight points they obtained $a_7(g_i) = 10\,080 \times 2^{-7}$.

VII. SERIES RESULTS

The simplest way to represent the series coefficients e_n and a_n [see Eqs. (14) and (24)] for any finite cluster or crystal lattice is in the form of Eqs. (77) and (78) together with Eqs. (80) and (81). The number of graphs which contribute to $e_1 - e_7$ are 1, 1, 2, 4, 7, 15, and 29, respectively, while the numbers of graphs that contribute to $a_1 - a_7$ are 1, 2, 4, 8, 16, 35, 82, respectively. The values of $e_n^{(m)}(g_i)$ and $a_n^{(m)}(g_i)$ for these graphs are given in Appendix D of Ref. 12.

For regular lattices, e.g., body-centered cubic and face-centered cubic lattices with $N \to \infty$, the lattice constants of connected graphs are proportional to N, and lattice constants of open graphs can be expressed in terms of the lattice constants of closed graphs and the coordination number of the lattice. We rewrite Eqs. (14) and (24) in the form

$$\ln Z/N = \ln Y + \sum_{n=2}^{\infty} e_n K^n, \tag{89}$$

and

$$\chi = \frac{C}{T}\left(1 + \sum_{n=1}^{\infty} a_n K^n\right), \tag{90}$$

where $C = N(g\mu)^2X/3k_B$. Note that the numbers $n!$ do not appear in these expressions and that e_1 has been set equal to zero by adjusting the zero of energy such that the internal energy is equal to zero at infinite temperature, that is, the constant Y^{-1} is subtracted from P_{ij} to make $\mathcal{H}$ traceless. The coefficients e_n and a_n will be written in the form

$$e_n = e_n^{(n)}Y^{-n} + e_n^{(n-2)}Y^{-n+2} + \cdots + e_n^{(1)}Y^{-1} \quad (\text{or } e_n^{(0)}), \tag{91}$$

$$a_n = a_n^{(n)}Y^{-n} + a_n^{(n-2)}Y^{-n+2} + \cdots + a_n^{(2)}Y^{-2} \quad (\text{or } a_n^{(1)}Y^{-1}). \tag{92}$$

The various quantities $e_n^{(m)}$ and $a_n^{(m)}$ are then given below:

$$e_2^{(0)} = \tfrac{1}{4}(\sigma+1), \quad e_2^{(2)} = -\tfrac{1}{4}(\sigma+1),$$

$$e_3^{(1)} = -\tfrac{1}{6}[(\sigma+1) - 6p_3],$$

$$e_3^{(3)} = \tfrac{1}{6}[(\sigma+1) - 6p_3], \quad e_4^{(0)} = -\tfrac{1}{24}[(\sigma+1)^2 - 12p_3],$$

$$e_4^{(2)} = \tfrac{1}{24}[(\sigma+1)(\sigma+4) - 84p_3 + 24p_4],$$

$$e_4^{(4)} = -\tfrac{1}{8}[(\sigma+1) - 24p_3 + 8p_4],$$

$$e_5^{(1)} = \tfrac{1}{60}[(\sigma+1)(5\sigma+4) - 15p_3(2\sigma+7) + 40p_4 + 30p_{5a}],$$

$$e_5^{(3)} = -\tfrac{1}{12}[(\sigma+1)(\sigma+2) - 3p_3(2\sigma+31) + 56p_4 - 12p_5 + 30p_{5a}],$$

$$e_5^{(5)} = \tfrac{1}{10}[(\sigma+1) - 60p_3 + 40p_4 - 10p_5 + 20p_{5a}],$$

$$e_6^{(0)} = \tfrac{1}{720}[(\sigma+1)(11\sigma^2+20\sigma+8) - 36p_3(7\sigma+4) + 72p_4 + 132p_{5a} + 96p_{6d}],$$

$$e_6^{(2)} = -\tfrac{1}{720}[(\sigma+1)(11\sigma^2+110\sigma+68) - 108p_3 \times (19\sigma+23) + 24p_4(20\sigma+93) - 600p_5 + 3132p_{5a} - 360p_{6a} - 360p_{6b} + 240p_{6c} + 1536p_{6d}],$$

$$e_6^{(4)} = \tfrac{1}{24}[(\sigma+1)(3\sigma+4) - 6p_3(10\sigma+51) + 8p_4 \times (2\sigma+39) - 140p_5 + 364p_{5a} + 24p_6 - 60p_{6a} - 60p_{6b} + 8p_{6c} + 192p_{6d}],$$

$$e_6^{(6)} = -\tfrac{1}{12}[(\sigma+1) - 114p_3 + 120p_4 - 60p_5 + 132p_{5a} + 12p_6 - 24p_{6a} - 24p_{6b} + 72p_{6d}],$$

$$e_7^{(1)} = -\tfrac{1}{5040}[(\sigma+1)(245\sigma^2+385\sigma+136) - 42p_3 \times (37\sigma^2+288\sigma+18) + 560p_4(5\sigma+6) - 910p_5 + 28p_{5a}(77\sigma+312) - 1260p_{6a} - 1456p_{6b} + 2352p_{6c} + 3864p_{6d} - 798p_{7c} - 840p_{7g} - 644p_{7h}],$$

$$e_7^{(3)} = \tfrac{1}{720}[(\sigma+1)(35\sigma^2+175\sigma+88) - 6p_3 \times (37\sigma^2+1128\sigma+348) + 80p_4(41\sigma+78) - 10p_5(60\sigma+343) + 52p_{5a}(29\sigma+234) + 720p_6 - 3060p_{6a} - 3808p_{6b} + 2256p_{6c} + 9192p_{6d} + 360p_{7a} + 360p_{7b} - 1314p_{7c} - 240p_{7e} + 360p_{7f} - 3000p_{7g} - 1532p_{7h}],$$

$$e_7^{(5)} = -\tfrac{1}{12}[2(\sigma+1)^2 - 3p_3(30\sigma+59) + 48p_4(\sigma+7) - 5p_5(2\sigma+47) + 10p_{5a}(2\sigma+59) + 84p_6 - 192p_{6a} - 216p_{6b} + 32p_{6c} + 576p_{6d} - 12p_7 + 30p_{7a} + 30p_{7b} - 68p_{7c} - 4p_{7e} + 30p_{7f} - 120p_{7g} - 96p_{7h}],$$

$$e_7^{(7)} = \tfrac{1}{14}[(\sigma+1) - 168p_3 + 280p_4 - 210p_5 + 476p_{5a} + 84p_6 - 168p_{6a} - 182p_{6b} + 504p_{6d} - 14p_7 + 28p_{7a} + 28p_{7b} - 56p_{7c} + 28p_{7f} - 84p_{7g} - 84p_{7h}], \tag{93}$$

and

$$a_1^{(1)} = (\sigma+1), \quad a_2^{(2)} = (\sigma+1)(\sigma-1),$$

$$a_3^{(1)} = -\tfrac{1}{3}[(\sigma+1) - 6p_3],$$

$$a_3^{(3)} = [(\sigma+1)(\sigma-1)^2 - 18p_3], \quad a_4^{(2)} = -\tfrac{1}{12} \times [(\sigma+1)(3\sigma-8) - 48p_3(\sigma-5) - 56p_4],$$

$$a_4^{(4)} = [(\sigma+1)(\sigma-1)^3 - 18p_3(2\sigma-5) - 32p_4],$$

$$a_5^{(1)} = \tfrac{1}{30}[(\sigma+1)(5\sigma+4) - 15p_3(2\sigma+7) + 40p_4 + 30p_{5a}],$$

$$a_5^{(3)} = -\tfrac{1}{6}[(\sigma+1)(\sigma^2-4\sigma+6) - 6p_3(6\sigma^2-34\sigma+85) - 8p_4(7\sigma-36) - 50p_5 + 204p_{5a}],$$

$$a_5^{(5)} = [(\sigma+1)(\sigma-1)^4 - 54p_3(\sigma^2-4\sigma+5) - 64p_4 \times (\sigma-3) - 50p_5 + 132p_{5a}],$$

$$a_6^{(2)} = \tfrac{1}{360}[(\sigma+1)(53\sigma^2-169\sigma-136) - 18p_3 \times (40\sigma^2-182\sigma-347) - 48p_4(3\sigma+115) + 1380p_5 + 24p_{5a}(30\sigma-347) + 756p_{6a} + 828p_{6b} - 216p_{6c} - 4944p_{6d}],$$

$$a_6^{(4)} = -\tfrac{1}{12}[(\sigma+1)(\sigma^3-6\sigma^2+14\sigma-16) - 12p_3 \times (8\sigma^3-52\sigma^2+138\sigma-219) - 8p_4 \times (21\sigma^2-124\sigma+344) - 20p_5(10\sigma-57) + 8p_{5a}(102\sigma-515) - 156p_6 + 636p_{6a} + 636p_{6b} + 64p_{6c} - 2544p_{6d}],$$

$$a_6^{(6)} = [(\sigma+1)(\sigma-1)^5 - 18p_3(4\sigma^3-21\sigma^2+42\sigma-34) - 32p_4(3\sigma^2-14\sigma+21) - 50p_5(2\sigma-7) + 24p_{5a}(11\sigma-40) - 72p_6 + 192p_{6a} + 188p_{6b} + 72p_{6c} - 624p_{6d}],$$

$$a_7^{(1)} = -\tfrac{1}{2520}[(\sigma+1)(245\sigma^2+385\sigma+136) - 42p_3 \times (37\sigma^2+228\sigma+18) + 560p_4(5\sigma+6) - 910p_5 + 28p_{5a}(77\sigma+312) - 1260p_{6a} - 1456p_{6b} + 2352p_{6c} + 3864p_{6d} - 798p_{7c} - 840p_{7g} - 644p_{7h}],$$

$$a_7^{(3)} = \tfrac{1}{360}[(\sigma+1)(46\sigma^3-205\sigma^2+347\sigma+264) - 6p_3(180\sigma^3-1029\sigma^2+3411\sigma+1292) - 4p_4(192\sigma^2-1076\sigma-7012) + 10p_5 \times (28\sigma-1495) + 4p_{5a}(270\sigma^2-1694\sigma+15589) + 2976p_6 + 12p_{6a}(126\sigma-1243) + 4p_{6b} \times (414\sigma-4531) - 16p_{6c}(27\sigma-406) - 48p_{6d} \times (206\sigma-1135) + 1388p_{7a} + 1454p_{7b} - 7820p_{7c} + 320p_{7d} - 348p_{7e} + 1292p_{7f} - 12540p_{7g} - 8272p_{7h}],$$

$$a_7^{(5)} = \tfrac{1}{6}[(\sigma+1)(\sigma^3-5\sigma^2+10\sigma-10) + 6p_3 \times (10\sigma^4-74\sigma^3+218\sigma^2-327\sigma+339) + 8p_4 \times (14\sigma^3-95\sigma^2+279\sigma-554) + 10p_5 \times (15\sigma^2-98\sigma+307) - 4p_{5a} \times (153\sigma^2-962\sigma+2399) + 12p_6(13\sigma-83) - 12p_{6a}(53\sigma-267) - 12p_{6b}(53\sigma-295) - 8p_{6c} \times (8\sigma-51) + 48p_{6d}(53\sigma-228) + 112p_7 - 456p_{7a} - 456p_{7b} + 1392p_{7c} - 96p_{7d} - 52p_{7e} - 456p_{7f} + 1992p_{7g} + 1796p_{7h}],$$

$$a_7^{(7)} = [(\sigma+1)(\sigma-1)^6 - 18p_3(5\sigma^4 - 32\sigma^3 + 84\sigma^2 - 110\sigma + 62) - 128p_4(\sigma^3 - 6\sigma^2 + 14\sigma - 14) - 50p_5 \times (3\sigma^2 - 16\sigma + 28) + 4p_{5a}(99\sigma^2 - 546\sigma + 971) - 144p_6(\sigma - 4) + 384p_{6a}(\sigma - 4) + 2p_{6b} \times (188\sigma - 777) + 144p_{6c}(\sigma - 4) - 1248p_{6d} \times (\sigma - 4) - 98p_7 + 256p_{7a} + 252p_{7b} - 580p_{7c} + 72p_{7d} + 96p_{7e} + 260p_{7f} - 588p_{7g} - 844p_{7h}]. \quad (94)$$

Here the p_{nx} are lattice constants for closed graphs per lattice site and $q = \sigma + 1$ is the coordination number of the lattice. The values p_{nx} and q for various crystal lattices are well known.[8] For convenience, the coefficients e_n for the cubic lattices and for several spin values are shown in Appendix A, and values of a_n for the cubic lattices and a number of two dimensional lattices are given in Appendix B. It is generally found that, similarly to the Heisenberg model,[11] the coefficients in these series expansions increase in smoothness as q increase. However, the series coefficients for the present model are much more irregular than those of the Heisenberg model, especially for large values of spin.

VIII. CHECKING PROCEDURES

Since there are numerous possibilities for errors to be made in the computations, it is important to be able to check the general expressions for the coefficients $e_n^{(m)}$ and $a_n^{(m)}$ before using them to estimate critical parameters. We have considered a large number of finite clusters of eight lines. For these clusters we can calculate the zero-field partition function and the low-field susceptibility series from Eqs. (93) and (94) by substituting the appropriate lattice constants. Comparing the results with those obtained from a direct machine calculation as described in Section VI yields full agreement up to e_7 and a_7 in all cases.

By observing the general expressions for $e_n^{(m)}$ and $a_n^{(m)}$ that we obtained [Eqs. (93) and (94)], we see that

$$\sum_m e_n^{(m)} = 0, \quad m = n, n-2, n-4, \cdots, 1 \text{ (or 0)}, \quad (95)$$

and

$$e_n^{(1)} = \tfrac{1}{2}a_n^{(1)}. \quad (96)$$

These two equations hold for all n and provide an additional check on our results. Equation (96) has been proved in Theorem 8, while Eq. (95) is a necessary consequence of the fact that, for $S = 0$, $\ln Z = 0$.

As a final check, we see that when we set S equal to $\frac{1}{2}$ and 1, respectively, our general results for e_n and a_n reduce exactly to those obtained previously by Domb and Wood[15] and by Allan and Betts,[16] respectively.

IX. ANALYSIS OF SERIES

A. Estimates of Curie Temperatures and Critical Indices

The Curie temperature T_C and the critical index γ for the susceptibility series are defined by

$$\chi \sim (T - T_C)^{-\gamma}, \quad \text{for } T \to T_C^+, \quad (97)$$

or

$$\sim (K_C - K)^{-\gamma}, \quad \text{for } K \to K_C^-, \quad (98)$$

where $K_C = J/k_B T_C$. For the face-centered cubic lattice T_C and γ were first estimated by means of the ratio method. From Eq. (98) the coefficients a_n in Eq. (90) have the property that for large n

$$\frac{a_n}{a_{n-1}} = \left(\frac{a_n}{a_{n-2}}\right)^{1/2} \to \frac{1}{K_C}\left(1 + \frac{\gamma - 1}{n}\right). \quad (99)$$

If we plot the two sets of values a_n/a_{n-1} and $(a_n/a_{n-2})^{1/2}$ vs $1/n$, each of the plots tends to a straight line as n increases and intersects with $1/n = 0$ at K_C^{-1} with slope $(\gamma - 1)K_C^{-1}$. If we plot $(a_n)^{1/n}$ vs $1/n$, the plot also intersects with $1/n = 0$ at K_C^{-1} but does not approach the intersection in a simple linear fashion for large n. Figure 4 illustrates these plots for the face-centered cubic lattice and for $S = 1$. The Curie temperature and the critical index can also be estimated by the method of Padé approximants.[17] The $[M,N]$ Padé approximant to a polynomial $f(z)$ is given by $P(z)/Q(z)$, where $P(z)$ and $Q(z)$ are polynomials of degree N and M, respectively, such that $Q(0) = 1$ and $f(z)$ agrees with the expansion of $P(z)/Q(z)$ for the first $M + N + 1$ terms.

If the divergence of χ at K_C is in the form shown in Eq. (98), then

$$(\chi)^{1/\gamma} \sim (K_C - K)^{-1}, \quad (100)$$

$$\frac{\chi'}{\chi} = \frac{d}{dK}\ln\chi = \frac{\gamma}{K_C - K}, \quad (101)$$

and

$$\frac{\chi''\chi}{(\chi')^2} = \frac{d}{dK}\left(\ln\frac{d\chi}{dK}\right) \Big/ \frac{d}{dK}(\ln\chi) = 1 + \frac{1}{\gamma}. \quad (102)$$

These equations suggest four methods of determining K and/or γ from χ:

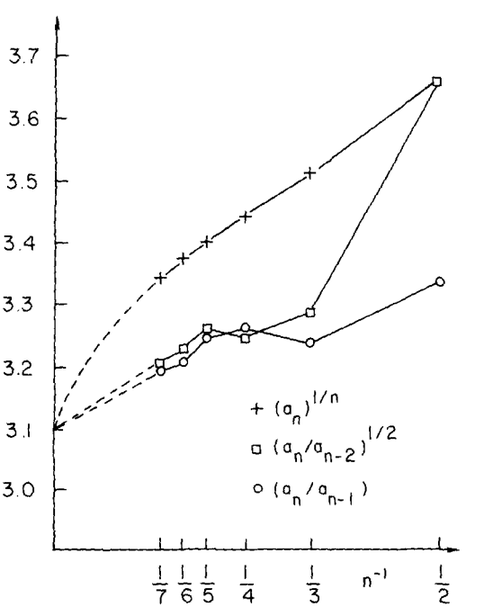

FIG. 4. Plots of $(a_n)^{1/n}$, $(a_n/a_{n-2})^{1/2}$, and $(a_n/a_{n-1})\ n^{-1}$ for the face-centered cubic lattice, $S = 1$.

(a) Choosing γ, K_C can be presented by appropriate poles of the Padé approximants to $(\chi)^{1/\gamma}$.

(b) Choosing K_C, γ can be obtained by evaluating Padé approximants to $(K_C - K)\chi'/\chi$ at $K = K_C$.

(c) For a Padé approximant to χ'/χ the appropriate pole gives K_C and the residue at this pole gives $-\gamma$.

(d) Evaluating Padé approximants to $\chi''\chi/(\chi')^2$ at $K = K_C$ gives $1 + 1/\gamma$.

For the face-centered cubic lattice and for $S = \frac{1}{2} - 3$ we have estimated K_C and γ by all the four methods. In method (a) instead of tabulating the various approximants in Padé table for several values of γ we have plotted the various approximants of K_C as functions of γ in the $K_C - \gamma$ plane. The proper values of γ and K_C are easily obtained from these curves in the region in which the various approximants coalesce. Figure 5 shows several of these curves for the face-centered cubic lattice and for $S = 1$. In method (b) the approximants of γ as functions of K_C are also plotted in the $K_C - \gamma$ plane. These curves are roughly parallel to and close to curves obtained from method (a) in the region of interest. The various approximants in methods (c) and (d) are rather irregular. However, they are not inconsistent with what methods (a) and (b) yield. The estimates of K_C and γ from Padé analysis are also in agreement with estimates based on the various ratio tests. The final estimates of γ and T_C are given in Figures 6 and 7, respectively. These results can be simply described by the equations

$$k_B T_C/J = 1.19 + 5.70Y^{-1} \tag{103}$$

and

$$\gamma = 0.48 + 2.16Y^{-1}. \tag{104}$$

Equations (103) and (104) hold for all S with the exception that $\gamma = 1.41 \pm 0.02$ for $S = \frac{1}{2}$.

For the body-centered cubic lattice and the simple cubic lattice the series coefficients are in general too irregular to estimate both K_C and γ either by ratio tests or by the Padé approximant method. However, within our precision, γ seems to be the same for all of the cubic lattices for each S. If the values of γ for the body-centered cubic and the simple cubic lattices are chosen to be the same as those of the face-centered cubic lattice, then an estimate of K_C from Padé approximants to $(\chi)^{1/\gamma}$ suggests that for all of the cubic lattices T_C can be described to within a few percent by

$$k_B T_C/J = 0.547(q - 1.6)(Y^{-1} + 0.21). \tag{105}$$

The estimates of T_C for the body-centered cubic and

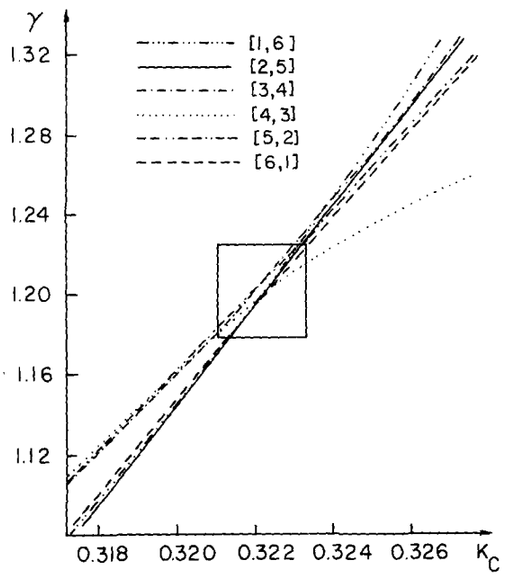

FIG. 5. Plots of several Padé approximants of K_C as functions of γ for the fcc lattice, $S = 1$.

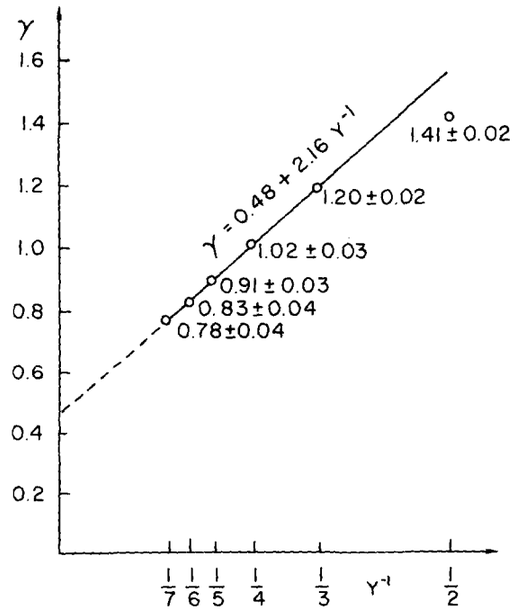

FIG. 6. Critical indices γ for the cubic lattices plotted vs Y^{-1}.

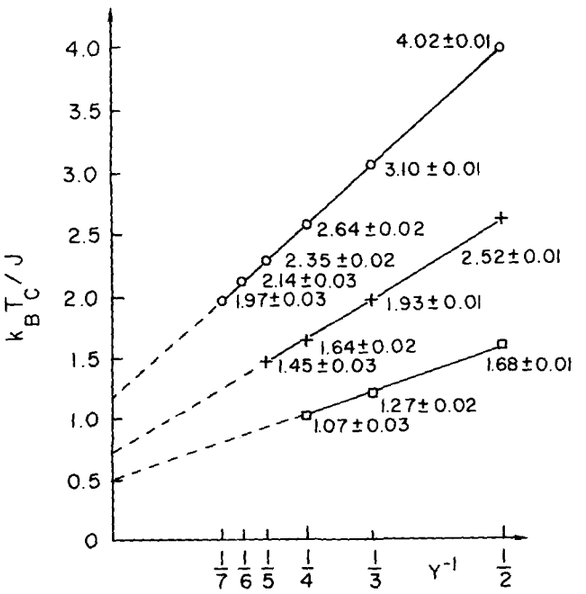

FIG. 7. Curie temperatures, $k_B T_C/J$, for the fcc lattice (denoted ○), b.c.c. lattice (denoted +), and sc lattice (denoted □) plotted vs Y^{-1}.

the simple cubic lattices are also contained in Figure 7.

For the two-dimensional lattices no consistent results could be obtained since the series are erratic and consist of positive and negative terms.

We have also investigated the specific heat series. The scatter in the various Padé approximants is too large to draw any conclusions concerning the critical temperature and the nature of the divergence of the specific heat at T_C. Considerably longer series would be needed for this purpose.

B. Critical Energy and Critical Entropy

Making use of the values of K_C in Eq. (105) the entropy series and the internal energy series obtained from Eq. (18) and (19) were also analyzed by the Padé approximant method. For the face-centered cubic lattice we found that

$$(S_\infty - S_C)/S_\infty = 0.494 - 0.353Y^{-1} \tag{106}$$

and

$$(E_\infty - E_C)/JN = 3.98 - 5.04Y^{-1}, \tag{107}$$

for all S with the exception that, for $S = \frac{1}{2}$, $(E_\infty - E_C)/JN = 1.60$. Here $S_\infty = Nk_B \ln Y$ and $E_\infty = 0$ are the entropy and the internal energy, respectively, at infinite temperature. For the body-centered cubic and the simple cubic lattices the convergence of the Padé approximants is in general fairly poor. However, the critical change of the entropy for the body-centered cubic and the simple cubic lattices seem also to vary linearly in Y^{-1} and for each spin value it is higher than that for the face-centered cubic lattice by an amount 0.025 and 0.055, respectively.

X. DISCUSSION AND CONCLUSIONS

Since there are $2S$ linearly independent isotropic interactions $(\mathbf{S}_i \cdot \mathbf{S}_j)^n [1 \leq n \leq 2S]$ for spin S particles, there are other kinds of order parameters[18,19] besides $\sum_i S_{zi}$, with which phase transitions might be associated. In order to study the possibility of such transitions we shall consider first the following modified Hamiltonian:

$$\mathcal{H}_n = \mathcal{H}_0 - \zeta Q_n, \tag{108}$$

where

$$\mathcal{H}_0 = -J \sum_{\langle ij \rangle} P_{ij} = -J\mathcal{P}, \tag{109}$$

$$Q_n = \sum_{i=1}^{N} S_{zi}^n, \quad n = 1, 2, \ldots 2S, \tag{110}$$

and ζ is some (fictitious) external magnetic field. We then define a generalized susceptibility χ_n by

$$\chi_n = \lim_{\zeta \to 0} \beta^{-1} \frac{\partial^2}{\partial \zeta^2} \ln \operatorname{tr} e^{-\beta \mathcal{H}_n}. \tag{111}$$

By definition $\chi_1 \sim \chi$. Since $\mathcal{P}$ and Q_n commute, as did $\mathcal{P}$ and Q, we have

$$\chi_n = \beta \Delta(Q_n). \tag{112}$$

In Ref. 20 the following theorem is proved.

Theorem 9:

$$\chi_n / \chi_1 = D_n(S) = (3/X)(W_{2n} - W_n^2). \tag{113}$$

Here $D_n(S)$ is a quantity which is independent of both *lattice* and *temperature* and W_n is given by Eq. (56). This means that for a given lattices, all of the $\chi_n \to \infty$ at the same T_C in exactly the same way.

Since the (dipolar) susceptibility (21) can also be written in the form

$$\chi = \tfrac{1}{3} g^2 \mu^2 \beta \sum_{i,j} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_\beta, \tag{114}$$

it is really necessary to investigate quantities of the form $\sum_{i,j} \langle (\mathbf{S}_i \cdot \mathbf{S}_j)^n \rangle_\beta$ for $1 \leq n \leq 2S$. However, for both computational and theoretical reasons, we have restricted our attention to the single quantity $\sum'_{i,j} \langle P_{ij} \rangle_\beta$. Here the prime in the double sum over i and j means that terms for which $i = j$ are to be excluded. The following theorem is also proved in Ref. 20.

Theorem 10:

$$\sum'_{i,j} \langle P_{ij} - (1/Y)\mathrm{I} \rangle_\beta = (4/Y) \sum'_{i,j} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_\beta. \tag{115}$$

Since $\langle \mathbf{S}_i^2 \rangle_\beta = X$, it immediately follows from this result and Eq. (114) that the left-hand side of Eq. (115) diverges at the same T_C and in exactly the same way as χ. Hence we must conclude that for the exchange interaction model the $2S$ "independent" multipolar phase transitions are in fact exactly degenerate with the dipolar transition. This means that the values of T_C and γ predicted on the basis of analysis of the high temperature susceptibility series are completely characteristic of this model.

Theorem 10 also suggests another approach for the derivation of the susceptibility series. Since $\sum'_{i,j} \langle P_{ij} \rangle_\beta$ is equal to χ except for a constant, χ can be obtained by calculating the quantities $\operatorname{tr} \sum'_{i,j} P_{ij} \mathcal{P}^n$ for each cluster. As was shown in Sec. IV, each Schrödinger exchange operator can be resolved into a direct sum of irreducible representations of the symmetric group. For a cluster of N sites it follows from Eqs. (46) and (49) that

$$\operatorname{tr} \sum_{i,j}' P_{ij} \mathcal{P}^n = \sum_\nu n_\nu [N(N-1) \chi_{1^{N-2}2}^{(\nu)} / \chi_{1^N}^{(\nu)}] \operatorname{tr} (\mathcal{P}^{(\nu)})^n, \tag{116}$$

where n_ν is given by Eq. (47) and $\chi_k^{(\Gamma)} = \operatorname{tr} P_k$ is given by Eq. (63). The labor required to derive χ by this approach is about the same as the previous approach which made use of Theorem 2.

The numerical results for the critical properties of the exchange interaction model have been shown in the previous section. Comparison of these results to those appropriate to the Heisenberg model[21,22] then shows that for $S > \frac{1}{2}$:

(1) Both T_C and γ for the present model are lower than those of the Heisenberg model.

(2) For the present model T_C is a decreasing function of spin, while T_C is an increasing function of spin for the Heisenberg model.

(3) For the exchange interaction model γ depends strongly on S and even becomes less than unity for large enough spin. On the other hand, for the Heisenberg model γ is a weak function of spin and is greater than 1.33 for all spin. Experimental values[23] of γ appear to lie in between the estimated values for these two models. Fisher[24] has recently suggested that the

observed value of the critical index γ_{obs} is related to the theoretical value by

$$\gamma_{obs} = \gamma/(1-\alpha), \tag{117}$$

where α is the theoretical value of the critical exponent of the specific heat series defined by

$$C_v \sim (T - T_C)^{-\alpha}, \quad \text{for } T \to T_C^+. \tag{118}$$

For $0 < \alpha < 1, \gamma_{obs} > \gamma$. This then shows that the theoretical estimates of γ when renormalized by the factor $(1-\alpha)^{-1}$ will be closer to the experimental values for the present model, while for the Heisenberg model the renormalized values of γ are in even further disagreement with the experimental values. This suggests that the present model may have more physical significance than originally thought.

(4) For the present model the fraction of the total entropy change occurring above T_C is higher than that of the Heisenberg model.

Hence we see that the inclusion of nonlinear terms in the Hamiltonian significantly affects the theoretical estimates of the critical parameters of magnetic systems.

The fact that T_C for the present model is lower than that of the Heisenberg model should be useful in a study of the possibility of a phase transition for the two-dimensional Heisenberg ferromagnet. If we can show that $T_C > 0$ for the two-dimensional exchange interaction model then there will be phase transitions for the two-dimensional Heisenberg model. However, as was previously mentioned, for large spin and/or for small coordination number, the series coefficients for the low-field susceptibility series are irregular and estimates of critical parameters from the high temperature series become quite inaccurate. It is then necessary that even more terms in the high temperature series be obtained.

APPENDIX A: VALUES OF e_n FOR SEVERAL CRYSTAL LATTICES

	$S=\frac{1}{2}$	$S=1$	$S=\frac{3}{2}$	$S=2$	$S=\frac{5}{2}$
Face-centered cubic lattice					
e_2	2.25	2.666 67	2.8125	2.88	2.916 67
e_3	2.25	1.777 78	1.406 25	1.152	0.972 22
e_4	0.468 75	−0.740 74	−1.259 76	−1.5168	−1.660 88
e_5	−1.3125	−1.481 48	−1.294 92	−1.107 45	−0.956 02
e_6	3.515 62	4.348 42	4.251 71	4.123 01	4.029 17
e_7	22.578 12	13.333 33	8.656 61	6.282 34	4.910 30
Body-centered cubic lattice					
e_2	1.5	1.777 78	1.875	1.92	1.944 44
e_3	−0.5	−0.395 06	−0.3125	−0.256	−0.216 05
e_4	0.4375	−1.086 42	−1.738 28	−2.0608	−2.241 52
e_5	0.375	2.304 53	2.378 91	2.159 62	1.919 24
e_6	0.531 25	1.285 78	3.688 96	5.259 01	6.241 93
e_7	−0.460 42	−6.285 14	−9.534 58	−9.961 30	−9.486 78
Simple cubic lattice					
e_2	1.125	1.333 33	1.406 25	1.44	1.458 33
e_3	−0.375	−0.296 29	−0.234 37	−0.192	−0.16204
e_4	−0.421 87	−0.962 96	−1.186 52	−1.296	−1.357 06
e_5	0.656 25	1.037 04	0.963 87	0.844 03	0.737 27
e_6	0.720 31	1.383 54	2.103 39	2.539 84	2.806 17
e_7	−1.579 69	−2.815 36	−3.148 86	−2.999 27	−2.739 34

APPENDIX B: VALUES OF a_n FOR SEVERAL CRYSTAL LATTICES

	$S=\frac{1}{2}$	$S=1$	$S=\frac{3}{2}$	$S=2$	$S=\frac{5}{2}$
Face-centered cubic lattice					
a_1	6.0	4.0	3.0	2.4	2.0
a_2	30.0	13.333 33	7.5	4.8	3.333 33
a_3	138.0	43.111 11	19.5	10.848	6.888 89
a_4	611.25	140.555 55	53.25	26.4336	15.472 22
a_5	2658.55	456.039 51	142.4	60.490 24	30.751 23
a_6	11 432.5125	1461.636 21	372.440 62	134.670 33	59.845 01
a_7	48 726.7262	4664.542 43	988.930 48	317.953 49	132.515 04
Body-centered cubic lattice					
a_1	4.0	2.666 67	2.0	1.6	1.333 33
a_2	12.0	5.333 33	3.0	1.92	1.333 33
a_3	34.666 67	9.777 78	3.833 33	1.770 66	0.888 89
a_4	95.833 33	21.851 85	8.208 33	4.043 73	2.351 85
a_5	262.7	45.195 06	16.4125	8.990 72	6.134 56
a_6	708.041 66	83.481 48	16.807 29	3.551 53	0.066 36
a_7	1893.289 68	168.439 35	24.685 73	−3.132 39	−10.696 58
Simple cubic lattice					
a_1	3.0	2.0	1.5	1.2	1.0
a_2	6.0	2.666 67	1.5	0.96	0.666 67
a_3	11.0	2.888 89	1.0	0.368	0.111 11
a_4	20.6250	4.722 22	1.781 25	0.880 80	0.513 89
a_5	39.025	7.451 85	3.340 63	2.208 64	1.712 04
a_6	68.777 08	7.190 53	0.618 09	−0.487 37	−0.647 19
a_7	119.429 76	8.197 00	−1.840 97	−3.868 18	−4.266 62

APPENDIX B: VALUES OF a_n FOR SEVERAL CRYSTAL LATTICES (Continued)

	$S=\frac{1}{2}$	$S=1$	$S=\frac{3}{2}$	$S=2$	$S=\frac{5}{2}$
Plane triangular					
a_1	3.0	2.0	1.5	1.2	1.0
a_2	6.0	2.666 66	1.5	0.96	0.666 67
a_3	8.5	2.888 89	1.4375	0.88	0.611 11
a_4	9.375	2.5	1.078 13	0.5928	0.375
a_5	11.025	1.624 69	0.098 44	−0.282 24	−0.386 73
a_6	16.964 58	2.069 14	0.209 89	−0.134 44	−0.192 21
a_7	21.152 68	4.766 14	2.472 38	1.771 23	1.428 05
Plane square					
a_1	2.0	1.333 33	1.0	0.8	0.666 67
a_2	2.0	0.888 89	0.5	0.32	0.222 22
a_3	1.333 33	0.148 15	−0.083 33	−0.138 67	−0.148 15
a_4	1.083 33	0.481 48	0.270 83	0.173 33	0.120 37
a_5	1.183 33	0.737 45	0.685 42	0.617 81	0.550 82
a_6	0.509 72	−0.677 02	−0.677 26	−0.053 45	−0.411 81
a_7	−4.821 83	−0.963 41	−1.125 63	−1.213 00	−1.190 45

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1 P. W. Anderson, Solid State Phys. **14**, 99 (1963); C. Herring, Magnetism IIB, 1 (1966).

2 G. A. T. Allan and D. D. Betts, Proc. Phys. Soc. (London) **91**, 341 (1967).

3 R. I. Joseph, Phys. Rev. **163**, 523 (1967).

4 E. Schrödinger, Proc. Roy. Irish Acad. **47**, 39 (1941).

5 G. A. Baker, H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Letters **20**, 146 (1966).

6 H. A. Kramers, Commun. Kamerlingh Onnes Lab. Leiden, Suppl. No. 83 (1936).

7 W. Opechowski, Physica **4**, 181 (1937); **6**, 1112 (1939).

8 C. Domb, Advan. Phys. **9**, 149 (1960).

9 The lattice constants defined here are the so-called high temperature lattice constant. See, for example, Ref. 8.

10 M. F. Sykes, J. W. Essam, B. R. Heap, and B. J. Hiley, J. Math. Phys. **7**, 1557 (1966).

11 G. S. Rushbrooke and P. J. Wood, Mol. Phys. **1**, 257 (1958).

12 H. H. Chen, thesis (The Johns Hopkins University, 1970). See Appendix A.

13 D. E. Littlewood, *The Theory of Group Characters* (Oxford at the Clarendon Press, Oxford, 1940).

14 T. Yamanouchi, Proc. Phys.-Math. Soc. Japan **19**, 436 (1937).

15 C. Domb and D. W. Wood, Proc. Phys. Soc. (London) **86**, 1 (1965).

16 For $S = 1$ and the fcc lattice we get $a_7 = 4664.5424$ while the results of Ref. 2 gives $a_7 = 4658.4738$. In private communication with Dr. Allan the mistake leading to the latter erroneous value has been discovered.

17 G. A. Baker, Advan. Theoret. Phys. **1**, 1 (1965).

18 M. Suzuki, Progr. Theoret. Phys. (Kyoto) **42**, 1086 (1969).

19 M. Blume and Y. Y. Hsieh, J. Appl. Phys. **40**, 1249 (1969) (abstract only).

20 H. H. Chen and R. I. Joseph, Phys. Rev. B **2**, 2706 (1970).

21 H. E. Stanley and T. A. Kaplan, J. Appl. Phys. **38**, 977 (1967).

22 M. E. Fisher, Rept. Progr. Phys. **30**, 615 (1967).

23 M. Vicentini-Missoni, R. I. Joseph, M. S. Green, and J. M. H. Levelt Sengers, Phys. Rev. B **1**, 2312 (1970).

24 M. E. Fisher, Phys. Rev. **176**, 257 (1968).

Canonical Dynamics of Spinning Particles in Gravitational and Electromagnetic Fields*

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In terms of the canonical form and the connection form on the bundle of Lorentz frames P over a space–time manifold V, a presymplectic form ω is defined on P, which induces a Poisson bracket on the set of real valued functions on the phase space of the system representing a spinning particle in an exterior gravitational and electromagnetic field. This structure coincides with the unique Poincaré invariant one for the free particle. Moreover, the projections into V of the integral manifolds of the kernel of ω on P yield precisely the world lines of a spinning particle as obtained for the dipole approximation of Dixon's equations of motion for extended test bodies in general relativity.

1. INTRODUCTION

It is well known that if a classical system can be described by a Lagrangian L, then not only the equations of motion are uniquely determined, but also a symplectic structure $\tilde{\omega}$ on the set of all motions M (equivalent to the phase space) of the system. The 2-form $\tilde{\omega}$ in turn induces a Lie algebra structure, the Poisson bracket, on the set of real valued functions on M (the "observables"). For several reasons it can be argued[1] that, apart from the equations of motion, this symplectic structure is all that is needed (for the comparison with quantized systems) and it seems considerably more fundamental than the Lagrangian itself. For example, the Lagrangian is not quite unique for given equations of motion and given $\tilde{\omega}$, and there are no Galilei or Poincaré invariant Lagrangians, but there are symplectic manifolds on which these groups act transitively. Moreover, to find a Lagrangian formulation it is necessary to separate phase space variables into position and momentum variables, a distinction which sometimes—as in the case of a particle with spin—looks slightly artificial.

One purpose of this paper is to illustrate, in the case of a test particle with spin in a curved space–time (a system described for fixed rest mass m and spin magnitude s at a given time t by initial data consisting of three position coordinates x^i, the 4-momentum p^α, and the 4-spin vector s^α, subject to the constraints $p^\alpha p_\alpha = -m^2$, $s^\alpha s_\alpha = s^2$, and $p^\alpha s_\alpha = 0$), that it may be easier to guess a suitable form for the symplectic